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DEVELOPMENT OF MULTIVARIATE ANALYSIS PROCEDURES FOR ONTARIO AIR QUALITY DATA

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DEVELOPMENT OF MULTIVARIATE ANALYSIS PROCEDURES FOR ONTARIO AIR QUALITY DATA

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INTRODUCTION

The Ministry of the Environment has a large air quality monitoring program in place that produces large quantities of high quality data on both local and regional scale problems. With support from the Ministry, we have examined several multivariate methods that can be used to extract useful information regarding the origins of airborne pollutants and their transport in the environment from these data sets and to provide these methods in the form of computer programs to the Air Resources Branch for their use. The results of the work supported by Grant 433G over the period of 1989 to 1992 are described in the this report.

THREE-WAY DATA ANALYSIS

Three-Mode Factor Analysis

In the studies conducted during the past three years, we have examined the use of three-mode factor analysis for the combined spatial and temporal analysis of environmental data. The concept of this approach is that environmental data are obtained at multiple locations at multiple times. Typically, eigenvector methods like principal components analysis and factor analysis have examined only one aspect of this system. However, a new method, three-mode factor analysis (Kroonenberg, 1983), offered the possibilities for simultaneously examining both the spatial and temporal covariance structure of a data set. We obtained the programs to perform these analyses from Dr. Kroonenberg and as part of our previous Ministry grant supported studies, we examined the utility of this method using two simulated data sets having known, well defined structures. The initial simulation study (Zeng and Hopke, 1989a) concluded that additional developmental work was needed to incorporate orthogonal rotations of the axes in order to arrive at a "simple structure" criterion similar to that used in ordinary factor analysis (Hopke *et al.*, 1976). This additional work was completed and the results of the entire study of three-mode factor analysis were presented by Zeng and Hopke (1990). The method was also applied to real world data with limited success (Zeng and Hopke, 1992a).

In the other studies of three-mode factor analysis, we tried to extend it to a quantitative source apportionment model as a three dimensional analog of target transformation factor analysis (TTFA) (Hopke, 1988; 1989). However, we found that even with error-free data, it was not possible to accurately reproduce the true simulation input values (Zeng, 1990). Thus, we began to consider alternative approaches to the analysis of three-way data.

Direct Trilinear Decomposition

Another approach to analyzing three way data has been developed in Professor Bruce Kowalski's group at the University of Washington. This method is called the Direct Trilinear Decomposition (DTD) (Sanchez and Kowalski, 1990). They developed the DTD method to process the data obtained in analytical chemistry with instruments that generate two-dimensional arrays of data for multiple samples. Their example was a three-way data set of emission-excitation fluorescence spectra for three mixtures of three organic compounds. With their DTD method, the emission and excitation spectra of all three compounds were predicted and the relative concentration of the compounds in the mixtures were calculated.

The following equation represents the trilinear model (Sanchez and Kowalski, 1990).

$$X_{ijk} = \sum_{n=1}^{N} a_n \otimes b_n \otimes c_n + E_{ijk} \qquad (1)$$

Note that X_{ijk} and E_{ijk} represent three-way matrices, and x_{ijk} and ϵ_{ijk} are their corresponding elements. X_{ijk} is the observed data matrix and E_{ijk} is the model error matrix. Vectors a_n, b_n, and c_n are *n*-th column vectors of matrices A, B, and C, and they represent the spectra or profiles of *n*-th components in the first, second, and third mode, respectively. The vector a_n is an *I* dimensional vector, b_n is *J*, and C_n is *K*. Equation (1) can be rewritten in scaler form:

$$x_{ijk} = \sum_{n=1}^{N} a_{in} b_{jn} c_{kn} + \epsilon_{ijk} \qquad (2)$$

The trilinear model is also called Parallel Factor Analysis (PARAFAC) or CANDECOMP model in the psychology literature (Kroonenberg, 1983; Harshman, 1970, 1976; Carroll and Chang, 1970).

The procedure for performing a trilinear decomposition based on the model in equation (1) is outlined in this subsection. A more detailed description and discussion and the algorithms can be found in Sanchez and Kowalski (1990). For the special case in which the three-way data block contains only two slices in mode 3 ($K=2$), a_n and b_n can be solved using the generalized rank annihilation method (Sanchez and Kowalski, 1988). In general, there are more than two slices ($K>2$). In order to use the generalized rank annihilation method, two linear combinations of all

slices need to be found. These two matrices may be generated using a two-component Tucker-1 model (Tucker, 1966; Kroonenberg, 1983):

$$X_{ijk} = \sum_{q=1}^{2} D_q \otimes u_q + E_{ijk} \tag{3}$$

Here the D_q are $I \times J$ matrices and the orthogonal vectors $\{u_q\}$ belong to the $\{c_n\}$ space.

$$D_q = \sum_{k=1}^{K} u_{qk} X_k \tag{4}$$

A suitable choice for the two $\{u_q\}$ vectors are the principal components of the unfolded X_{ijk} in the $\{c_n\}$ space.

On another hand, a slice of the data (e.g., *k*-th slice) can be expressed as the following expression according to the model (equation 1):

$$X_k = \sum_{n=1}^{N} c_{kn} a_n \otimes b_n \tag{5}$$

Combining equations 4 and 5:

$$D_q = \sum_{k=1}^{K} u_{kq} \sum_{n=1}^{N} c_{nk} a_n \otimes b_n = \sum_{n=1}^{N} (a_n \otimes b_n) \sum_{k=1}^{K} c_{nk} u_{kq} \tag{6}$$

Defining

$$v_{nq} \equiv \sum_{k=1}^{K} c_{kn} u_{qk} = c_n \cdot u_q \tag{7}$$

Then for D_1 and D_2:

$$D_1 = \sum_{n=1}^{N} v_{n1} a_n \otimes b_n$$
$$D_2 = \sum_{n=1}^{N} v_{n2} a_n \otimes b_n \tag{8}$$

Now generalized rank annihilation can be applied to these two matrices. If the $\{b_n\}$ vectors are linearly independent, and $b_n \cdot b_q = \delta_{nq}$, the Kroneker delta ($n=q \Rightarrow \delta_{nq}=1$, $n \neq q \Rightarrow \delta_{nq}=0$), then

$$\begin{aligned} D_1 b_n &= v_{n1} a_n \\ D_2 b_n &= v_{n2} a_n \end{aligned} \tag{9}$$

Eliminating a_n from the equations, the generalized eigenvalue-eigenvector problem is obtained (Sanchez and Kowalski, 1986):

$$D_1 b_n v_{n2} = D_2 b_n v_{n1} \tag{10}$$

where $\{b_n\}$ vectors are eigenvectors. Equation 10 can be solved by projecting D_1 and D_2 to square matrices and then applying the QZ algorithm for simultaneous diagonalization (Sanchez and Kowalski, 1986;1988).

After $\{b_n\}$ vectors are found, the $\{a_n\}$ vectors can be evaluated by

$$a_n = \frac{D_1 b_n}{\|D_1 b_n\|} \tag{11}$$

Finally, the vectors $\{c_n\}$ can be estimated by least squares from $\{a_n\}$ and $\{b_n\}$:

$$P: \quad p_{kn} = \sum_{i=1}^{I} \sum_{j=1}^{J} x_{ijk} a_{in} b_{jn}$$

$$Q: \quad q_{st} = \sum_{i=1}^{I} (a_{is} a_{it}) \sum_{j=1}^{J} (b_{js} b_{jt}) \qquad (12)$$

$$C = PQ^{-1}$$

The key to applying DTD in receptor modeling is to establish the relationship between the terms in the model [equation 1] and the physical properties of the airshed. If the three-way data matrix is arranged such that mode 1 (i) corresponds to the chemical species, mode 2 (j) to sampling time period, and mode 3 (k) to site, the N vectors $\{a_n\}$ can represent the source profiles. Two new concepts are introduced so that the model can be fitted into the physical system. "Emission pattern" is used to reflect the relative variations of source emission strength changing with time at the receptors. Like a source profile, the emission pattern can be considered to be another property of a source. Each source has its own emission pattern. With the source profile and emission pattern, a source can be characterized more completely and accurately. The vectors $\{b_n\}$ in model then correspond to the emission patterns.

Another concept, the site coefficient, is introduced to refer to the vectors $\{c_n\}$ in equation 1. The site coefficients indicate the variation in source strength from site to site. With these definitions, a three-way air quality data set could be decomposed into source profiles (a_n), emission patterns (b_n), and site coefficients (c_n). However, each of these profiles is a relative profile, and its values may not agree with actual physical scales.

The ultimate goal of receptor modeling is not only to identify sources, but also to apportion the sample mass among the sources. In order to estimate the mass contributions, a matrix reconstruction procedure is introduced after DTD. The mass contribution of source n at site k in period j, $\bar{z}_{njk}$, can be obtained by combining the emission pattern and the site coefficient.

$$\bar{z}_{njk} = b_{jn} c_{kn} \qquad (13)$$

If the source profile $\bar{y}_{in}$ is defined as

$$\bar{y}_{in} = a_{in}, \tag{14}$$

the model in scaler form becomes:

$$x_{ijk} = \sum_{n=1}^{N} a_{in} b_{jn} c_{kn} = \sum_{n=1}^{N} \bar{y}_{in} \bar{z}_{njk} \tag{15}$$

The scale of $\bar{y}_{in}$ and $\bar{z}_{njk}$ do not necessarily agree with the actual scale of source profiles and mass contributions. Similar to TTFA (Hopke, 1988; 1989), a regression procedure can be used to find a set of scaling coefficients, s=$\{s_n\}$, such that

$$x_{ijk} = \sum_{n=1}^{N} \left(\frac{\bar{y}_{in}}{s_n} \right) (s_n \bar{z}_{njk}) = \sum_{n=1}^{N} y_{in} z_{njk} \tag{16}$$

where y_{in} and z_{njk} are the scaled source profiles and contributions. The regression is based on

$$mass_{jk} = \sum_{n=1}^{N} z_{njk} = \sum_{n=1}^{N} s_n \bar{z}_{njk} \tag{17}$$

Figure 1 illustrates the overall DTD procedure to provide a quantitative source resolution. The application of DTD in receptor modeling was examined in methodological studies using simulated data sets and a small real data set. Exact reproduction of error-free data was obtained and quite good reproduction was found even for realistic levels of error in the data. The results of these studies are presented by Zeng and Hopke (1992b). We are currently pursuing the use of this method with the APIOS particulate data and those studies will be the subject of a future report.

POTENTIAL SOURCE CONTRIBUTION FUNCTION

We have explored another approach to data analysis based on combining meteorology with chemical composition data. The method was presented by Malm *et al.* (1986) in their analysis of particulate composition data from samples obtained at the Grand Canyon. They were able to relate a variety of different sources types to probable locations in the western United States. We have applied these methods to precipitation chemistry data for 1984 to 1986 from the Dorset, Charleston Lake, and

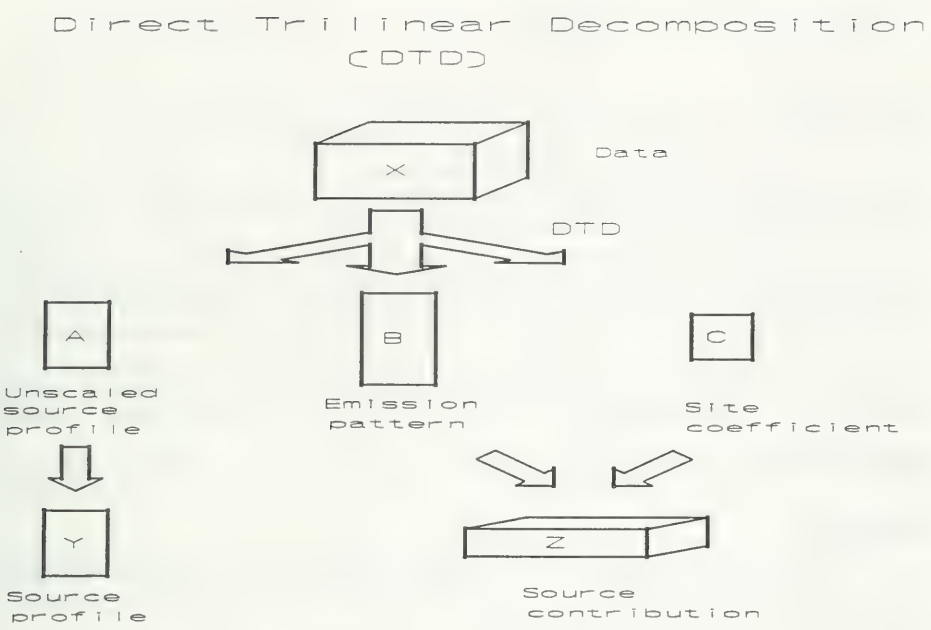

Figure 1. Outline of DTD model.

Longwoods sites (Zeng and Hopke, 1989b). We have applied potential source contribution function (PSCF) analysis to particulate data from Dorset using the trajectories calculated by D. Yap based on surface meteorology (Zeng and Hopke, 1992c). We have also examined the PSCF results for the trace elemental data for particles and precipitation samples obtained at Dorset from 1986 to 1988 (Hopke and Gao, 1992).

There has been concern raised in discussions of our work that there could be problems because we have used only surface meteorology based trajectories and only looked backward in time for a period of 48 hours. Thus, we asked Dr. M. Olson of the Atmospheric Environment Service (AES) to calculate trajectories at multiple heights in the atmosphere (1000, 925, and 850 hPa) using the AES three dimensional trajectory code (Olson *et al.*, 1978). On the basis of these trajectories, we have calculated PSCF fields at each of the three pressure heights. In addition we have examined the combination of both the two and three heights by calculating the Total Potential Source Contribution Function (Cheng *et al.*, 1992). We have found that the incorporation of the two lower levels (1000 and 925 hPa) provides a good representation of the known emission sources for SO_2 while the

inclusion of the 850 hPa suggested areas that are too far from Dorset to be significant source regions.

Other studies of the utility of this method as a function of the distance scale of the transport have also been performed. A study of the transport of gaseous and ionic species measured during the Southern California Air Quality Study (Gao *et al.*, 1992) was performed to examine the utility of PSCF at a sub-regional scale. In this study, the trajectories are resolved on a 5 km x 5 km grid cell size and individual sources can be identified through their presence in particular high potential cells. The utility of PSCF analysis at various levels of scale length for the atmospheric transport has been reviewed by Hopke *et al.* (1992) where it appears that the method can provide useful information over the range of scale from urban to semi-global assuming that adequate back-trajectory information is available. It is anticipated that PSCF can be applied to the identification of source locations of other atmospheric species such as hazardous organic compounds. Such work will be pursued during the next grant period.

SUMMARY

The primary objectives during the past three years were to develop three way data analysis as a quantitative tool for receptor modeling. The initial results have been encouraging and the further analyses of Ministry data is now in progress. We have also continued our development and study of the Potential Source Contribution Function by incorporating multiple height data into the analysis and examining the location resolution as a function of transport scale. Again useful results have been obtained for Ministry data as well as data from other locations and additional applications of the method can be anticipated.

PERSONNEL

This project was directed by Philip K. Hopke, Robert A. Plane Professor of Chemistry at Clarkson University in Potsdam, N.Y. It has supported the Ph.D. thesis work of Yousheng Zeng (Ph.D., 1990) and Ning Gao who joined Professor Hopke's group in January 1991 and is expected to complete her studies during 1993. During part of 1992, the grant supported a portion of the effort of Dr. M.D. Cheng, a research associate in Professor Hopke's receptor modeling group.

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